EPJ manuscript No.
(will be inserted by the editor)

On the existence of stationary states during granular compaction

Philippe Ribière[1], Patrick Richard[1], Pierre Philippe[2], Daniel Bideau[1] and Renaud Delannay[1]

[1] Groupe Matière Condensée et Matériaux, UMR CNRS 6626, Université de Rennes 1, Campus de Beaulieu, F-35042 Rennes cedex, France
[2] Cemagref, Aix-en-Provence, Le Tholonet BP 31, F-13612 Aix-en-Provence Cedex 1, France

Abstract. When submitted to gentle mechanical taps a granular packing slowly compacts until it reaches a stationary state that depends on the tap characteristics. The properties of such stationary states are experimentally investigated. The influence of the initial state, taps properties and tapping protocol are studied. The compactivity of the packings is determinated. Our results strongly support the idea that the stationary states are genuine thermodynamic states.

PACS. 45.70.-n Granular systems. – 45.70.Cc Static sandpiles; granular compaction – 81.05.Rm Porous materials; granular materials

1 Introduction

In the absence of an external drive, granular materials rapidly come to rest. This is a consequence of their dissipative interactions and of the irrelevance of thermal energy (in this case thermal energy is negligible compared to the energy needed to move a grain). Thus standard thermodynamics is not applicable to those systems. Edwards and Oakeshott [1] proposed a non-standard thermodynamic description for static granular media. This theory postulates that a granular packing at rest can be described by suitable ensemble averages over its blocked "jammed" states. If one assumes that all the available blocked states have the same probability to occur and that the volume is analog to the energy of thermal systems, the configurational entropy of a granular packing is $S = \lambda \ln \Omega(V, N)$, where λ is the equivalent of the Boltzmann constant and $\Omega(V, N)$ the number of mechanically stable configurations of N particles in the volume V. A temperature-like state variable, the compactivity $X = \partial V/\partial S$ can then be introduced. This theory was partially investigated by recent experiments [2,3,4,5], numerical simulations of models [6, 7,8,9,10]. These experiments have established that a granular system subjected to a tapping dynamics slowly compacts (i.e. the packing fraction increases) and reaches a stationary state that depends on the tapping intensity. The existence of a stationary state is the first essential step to justify the validity of a thermodynamic-like description for granular packings.
Here we experimentally study the dynamics of granular media submitted to gentle mechanical taps. We mainly focus on the properties of the stationary states and discuss the validity of such configurations as genuine thermodynamic states. Using the fluctuations of the packing fraction at stationary state we will determine the compactivity introduced by Edwards and Oakeshott [1] and compare our results with those obtained for granular packings submitted to fluid flow pulses [11].
The outline of this paper is the following. We first describe the experimental setup. Then, in section 3, we rapidly recall the previous results obtained on the relaxation of the packing fraction during compaction. Section 4 is devoted to the influence of the initial conditions on the stationary state and section 5 reports the dependence of our results on the frequency used to generate the taps. The cases of annealing and memory effects are studied in section 6. Finally, section 7 is devoted to the conclusions of this work.

2 Experimental setup

The experiments (Fig. 1a) are performed with $d = 1$ mm diameter glass spheres placed in a glass cylinder of diameter $D = 10$ cm. The cylinder containing the grains is tapped vertically at regular intervals ($\Delta t = 1$ s). Each tap is controlled by an entire cycle of a sine wave at a fixed frequency $f = 30$ Hz: $V(t) = V_{MAX}(1 - \cos(2\pi f t))/2$ for $0 < t < 1/f$ and $V(t) = 0$ elsewhere (Fig. 1b). This applied voltage is connected to an electromagnetic exciter (LDS V404) which induces a vertical displacement to a moving part supporting the container and the beads. The resulting motion of the whole system is monitored by an accelerometer at the bottom of the container. This motion is more complicated than a simple sine wave: at first the system undergoes a positive acceleration followed by a negative acceleration with a minimum equal to $-\gamma_{max}$. After the applied voltage stops, the system relaxes to its normal position (Fig. 1c). When γ_{max} is large enough, the bead packing takes off from the bottom of the container and achieves a flight until it crashes back to the bottom.

This crash is visible on the signal of the accelerometer: a negative acceleration $-\gamma_{fall}$ corresponding to the fall of the grains is clearly visible. The control parameters are the

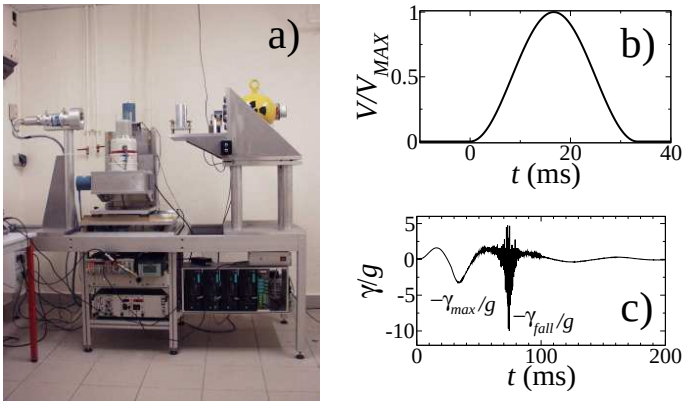

Fig. 1. a) Picture of the experimental setup, b) the signal controlling the tap (here $f = 30$ Hz) and c) the acceleration monitored by the accelerometer (here $\Gamma \approx 3.3$).

frequency f used to generate the tap and the tapping intensity $\Gamma = \gamma_{max}/g$, where $g = 9.81$ m.s^{-2}. The packing fraction is measured using a γ-ray absorption setup [3]. The measure is deduced from the transmission ratio of the horizontal collimated γ beam through the packing: $T = A/A_0$, where A and A_0 are, respectively, the activities counted on the detector with and without the presence of the granular medium. From Beer-Lambert law for absorption, we can derive an estimation of the volume fraction in the probe zone: $\rho \approx -(\mu D)^{-1} \ln(T)$, where μ is the absorption coefficient of the beads. It was evaluated experimentally to $\mu \approx 0.188$ cm^{-1} for our γ beam of energy 662 keV (^{137}Cs source). The collimated γ beam is nearly cylindrical with a diameter of 10 mm and intercepts perpendicularly the vertical axis of the vessel. In order to reduce the relative fluctuations due to number of emitted photons we use an acquisition time of 300 seconds for each measure which leads to a precision of 0.001. The packing fraction of the sample is estimated from the ratio T averaged on approximately 7 cm height from the bottom of the cylinder.

3 Packing fraction relaxation laws

The first quantity of interest in compaction is the packing fraction (or density) ρ, defined as the ratio of the volume of the grains to the total volume occupied by the packing. A few characteristic values of ρ for mono-sized sphere packings have to be reminded. The maximal packing fraction reached in a random packing of spheres (the so-called random close packing fraction) is $\rho_{RCP} \approx 0.64$. This value is significantly lower than the maximal packing fraction obtained for face-centered-cubic (or hexagonal compact) packing ($\rho_{max} \approx 0.74$). Another limit is the so-called random loose packing corresponding to the less dense mechanically stable packing ($\rho_{RLP} \approx 0.58$).

In a pioneering paper, Knight et al. [2] in Chicago first considered packing fraction relaxation law in granular compaction. They showed that, starting from a loose packing of beads confined in a very narrow and tall tube (diameter 1.88 cm and 87 cm high, bead diameter 2 mm), a succession of vertical taps induces a progressive and very slow compaction of the system. They found that the relaxation law can be well fitted by an inverse-logarithmic law, the so-called Chicago fit:

$$\rho(t) = \rho_\infty - \frac{\rho_0 - \rho_\infty}{1 + B \ln(1 + t/\tau)}.$$

For a given frequency f, the fitting parameters ρ_∞, ρ_0, B and τ essentially depend on Γ. The small number of grains in a tube diameter (≈ 10) allows for a local measurement of the packing fraction with a capacitive method and prevents any convection in the packing. Nevertheless it induces strong boundary effects that may be responsible for crystallization (some packing fraction values obtained are well above the random close packing limit) visible in some of the experiments reported in [12] and in the values of ρ_∞ obtained in [2].

More recently Philippe and Bideau [3] carried out new compaction experiments using the setup described in section 2 with about 100 grains in the tube diameter. This restricts the boundary effects but, contrary to the Chicago group's experiments, allows convection. The relaxation law obtained by these authors differs significantly from those obtained by Knight et al. [2], especially for the long-time behavior. Indeed whereas in previous experiments no clear evidence of convergence to a steady state has been established, such an evidence in definitely produced by our experiments and may correspond to a dynamical balance between convection and compaction. The relaxation is better fitted by the Kohlrausch Williams Watts law (KWW law) - a stretched exponential - :

$$\rho(t) = \rho_\infty - (\rho_\infty - \rho_0) \exp\left[-(t/\tau)^\beta\right]$$

where ρ_∞ and ρ_0 correspond respectively to the steady state and to the initial packing fraction value (figure 2). The adjustable parameters τ and β are here respectively the relaxation time and a parameter related to the stretching of the exponential. This characteristic time scale is found to be well described by an Arrhenius behavior

$$\tau = \tau_A \exp\left[\frac{\Gamma_A}{\Gamma}\right]. \quad (1)$$

Such kind of relaxation law is also found for strong glasses (the dimensionless acceleration Γ plays the role of the temperature). Note that, Lumay and Vandewalle recently recover KWW-like laws for compaction of 2D granular packings [13]. As can be seen in figure 2, a stationary state is reached (if the number of taps is large enough). Figure 3 reports the evolution of the packing fraction of the stationary state as a function of the tapping intensity Γ for $f = 30$ Hz. We observe a decrease of the packing fraction with Γ. To reach an important value of packing fraction it is thus essential to use a small tapping intensity.

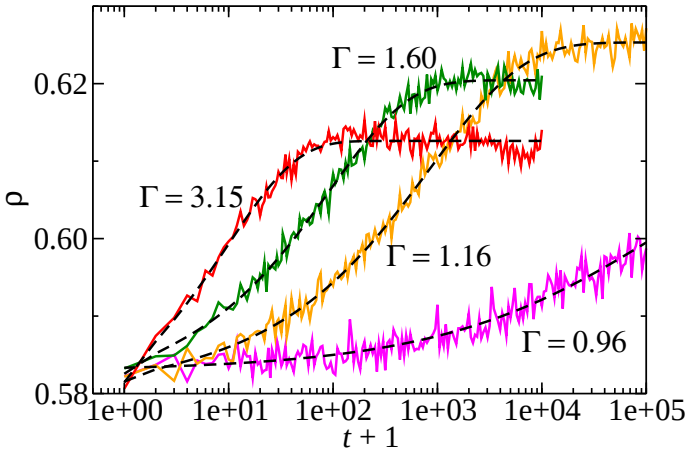

Fig. 2. Packing fraction versus the number of taps in our experiments. The curves are KWW fits (see text). The tap frequency is $f = 30$ Hz.

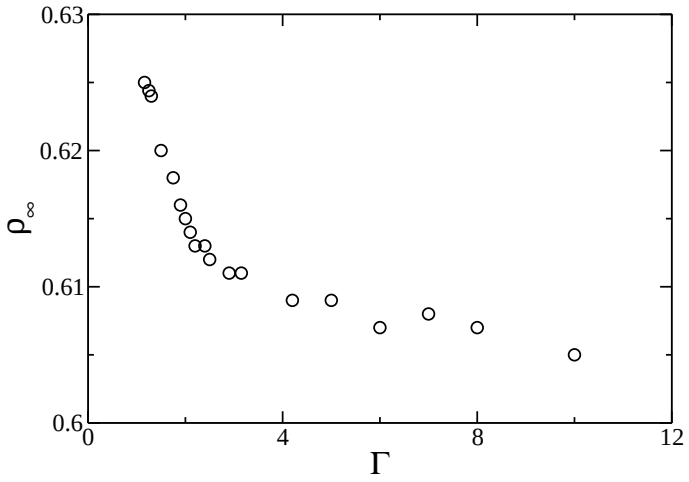

Fig. 3. Packing fraction of the stationary state versus the tapping intensity for $f = 30$ Hz

4 Influence on the initial state

The question of the existence of a stationary state during granular compaction is essential to validate any thermodynamic approach. As reported in the previous section, loose packings submitted to gentle mechanical taps continuously compact until they reach a stationary state. Does this state depend on the initial conditions ? To answer this question, we submit two granular packings of different packing fractions (one dense with a packing fraction close to that of the random close packing : packing 1 and one loose with a packing fraction close to that of the random loose packing : packing 2) to taps of the same intensity. As reported in figure 4a (see also [14]) after a transient the two packings reach the same value of packing fraction. This behavior is observed for a large range of Γ (figure 4b). This shows that, for a given tapping process, the value of the packing fraction at stationary state does not depend on the initial conditions. For the two initial packings considered, the number of taps needed to reach

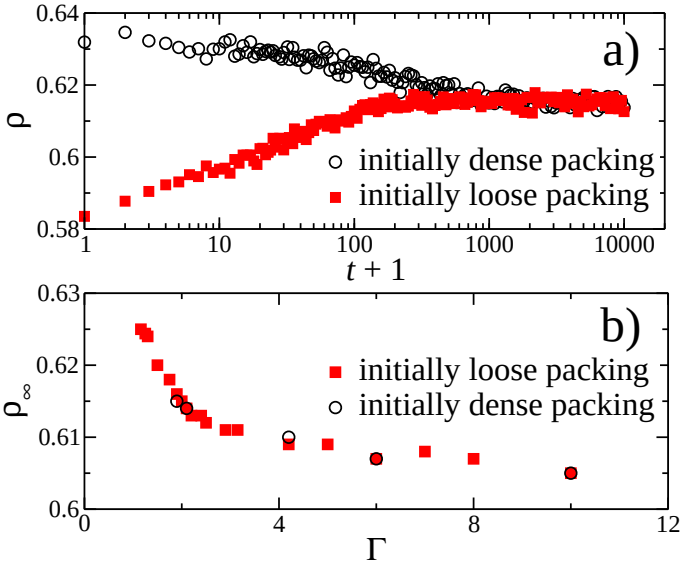

Fig. 4. a) Evolution of the packing fraction versus the number of taps for an initially dense packing and an initially loose packing. We observe that the final packing fraction value is similar for the two packings ($\Gamma = 2.1$ and $f = 30$ Hz). b) Evolution of the final packing fraction versus the tapping intensity for an initially dense packing and an initially loose packing.

the steady state is larger for decompaction than for compaction. This is true for any value of Γ [14]. This can be intuitively understood : contrary to decompaction, compaction is facilitated by the gravity.

5 Influence of the tap duration

As mentioned in previous section, the stationary state does not depend on the initial conditions. We can modify the frequency generating the tap and study its effect on the stationary state. We report in Fig 5a the stationary values of the packing fraction ρ_∞ as well as its fluctuations $\Delta\rho_\infty$ recorded after a sequence of taps of duration $1/f$ (where f is 30 Hz, 60 Hz or 90 Hz) and intensity Γ.

Even though ρ_∞ and its fluctuations at stationary state depend more or less on both Γ and $1/f$ (for the latter it is particularly visible at low values of Γ), when $\Delta\rho_\infty$ is plotted as a function of ρ_∞ the data collapse, within errors bars, onto a single master curve (figure 5b). This shows that ρ_∞ and $\Delta\rho_\infty$ are linked. Note that the smallest values of $\Delta\rho_\infty$ might be perturbated by the fluctuations of the γ-ray source. In other words, no matter how the state with packing fraction ρ_∞ is attained. This result is in agreement with those obtained very recently by Pica Ciamarra et al. [15] with numerical simulations of granular packing submitted to fluid flow pulses. These results strongly support the idea that such stationary state are indeed genuine "thermodynamic states". Moreover, in [15], the authors demonstrate that at stationarity a granular packing can

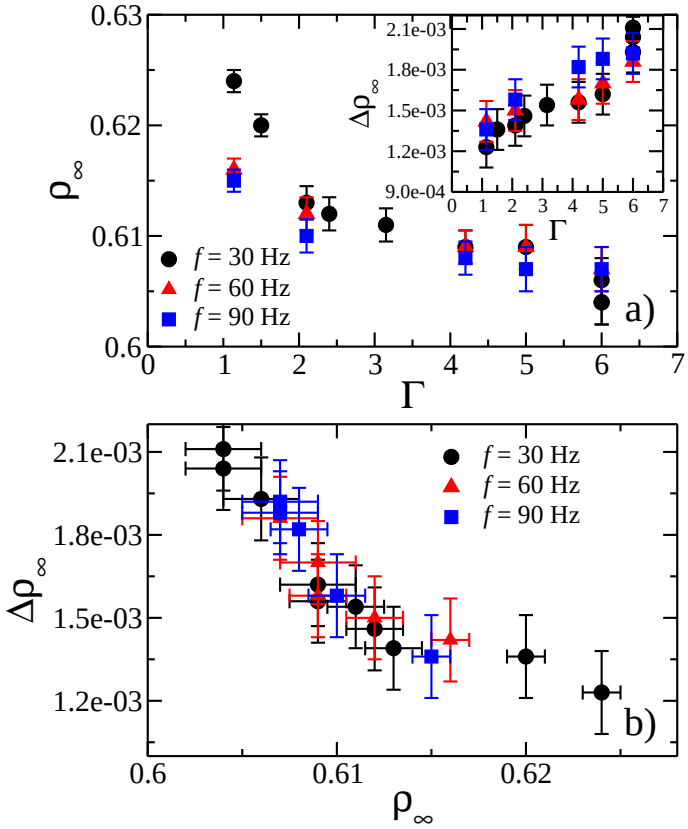

Fig. 5. a) The main panel shows the packing fraction obtained at stationarity as a function of the tapping acceleration for different values of the frequency of the mechanical tapping. The inset shows the dependence of the packing fraction fluctuations on Γ and f. b) Plot of the packing fraction fluctuations versus the packing fraction at stationarity. All the data collapse on a single master curve, showing that $\Delta\rho_\infty$ and ρ_∞ are linked and that this link does not depend on the values of f and Γ.

be described by only one parameter, the packing fraction, all the other observables being characterized by this parameter. The strong resemblance of our results with the ones reported in [15] suggests that this is also true for experiments of granular packings submitted to mechanical tapping.

It should be pointed out that this is true only at the stationary state. Indeed, during the transient, memory effects can be observed [16]. In this case, the packing fraction is in general no more enough to fully characterize the packing and other observables are needed [17]. Knowing the dependency of $\Delta\rho_\infty$ on ρ_∞ enables us to determine Edwards' compactivity, which the equivalent of the temperature in thermal systems. Using the fluctuation dissipation theorem [18] we can derive

$$\frac{\lambda\rho_g}{m}X(\rho) = \left(\int_{\rho_{RLP}}^{\rho}\left(\frac{\varphi}{\Delta\varphi}\right)^2 d\varphi\right)^{-1}$$

where we have used $X(\rho_{RLP}) = \infty$. In this expression m is the grain mass and ρ_g the grain density. We analytically solve this equation using for $\Delta\rho_\infty(\rho_\infty)$ a 2^{nd} order polynomial fit (figure 6). In order to compare our results with those reported in [11] we use the same value for the packing fraction of the random loose packing : $\rho_{RLP} = 0.573$. We observe a decrease of X with ρ_∞. al-

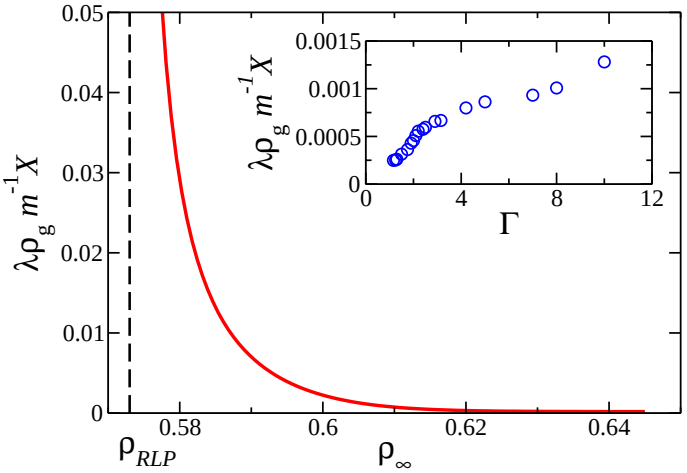

Fig. 6. Compactivity as a function of the average packing fraction at stationary state. (Inset) Evolution of the compactivity versus the tapping intensity for $f = 30$ Hz.

though the evolution of $\Delta\rho_\infty$ is different, the behavior of $X(\rho_\infty)$ is similar to that found for granular packings submitted to fluid flow tapping [11]. We also report in the inset of figure 6 the evolution of the the compactivity versus the tapping intensity Γ for $f = 30$ Hz. We observed that these two quantities are linked: the temperature-like compactivity increases with the tapping intensity. Note that the change of behavior observed around $\Gamma = 2$ may be explained by the two different convective regimes observed in our system [4]. In the analogy between glassy systems and granular packings undergoing compaction, Γ is supposed to play the role of temperature at the stationary state. So the above-mentioned result reinforce this analogy. It should be interesting to compare the compactivity with other "temperatures" defined for granular media as the granular temperature (defined as the velocity fluctuations) or the effective temperature defined through the out-of-equilibrium fluctuation-dissipation theorem [19].

6 Annealing and memory effects

Further insights into the understanding of the stationary state characteristics can be gained by allowing the tap intensity to vary in time. Annealing experiments phenomena in granular media undergoing compaction have been reported previously in [12]. The authors proceeded as follows: starting from a loose packing of grains, the material is tapped at a given intensity Γ for a given time t (10^5 taps). Γ is then modified and the compaction process continued for t taps (see figure 7a). The increase of Γ corresponds to an increase of the average packing fraction except for values larger than three for which a slow

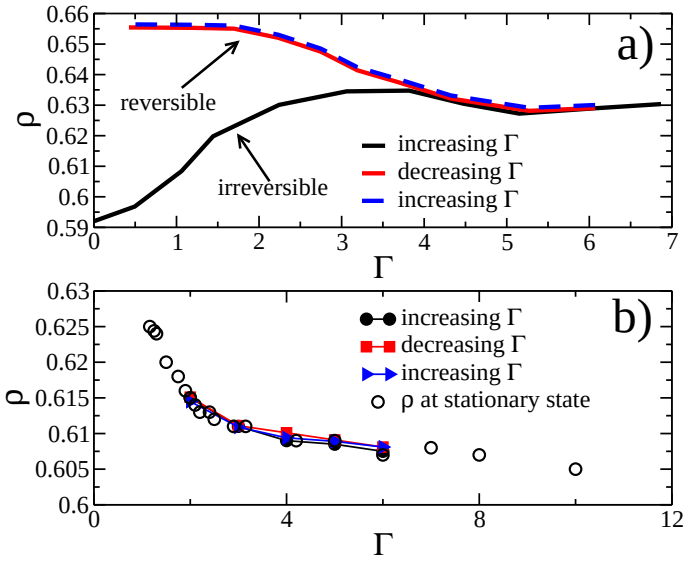

Fig. 7. a) Sketch of the evolution of the packing fraction during annealing experiments reported in [12]. b) Results of annealing experiments in our experiments with $f = 30$ Hz. No aging is observed.

decrease can be observed. If Γ is then reduced, the packing fraction, rather than following reversely the previous curve, still increases. This new curve, contrary to the previous is reversible. Using our setup and the same protocol (with $f = 30$ Hz), we only recover the reversible branch (see figure 7b). This can be explained by the fact that the number of taps used is large enough to allow our system to reach stationarity. Thus, aging and irreversible-reversible behaviors are observed only when the stationary state is not reached. Note that these experiments also confirm that the stationary state only depends on the tapping protocol and not on the initial conditions.

We also carried out memory effects experiments. Let us recall that Josserand et al. [16] carried out experiments on the response function of a granular media undergoing compaction to a sudden perturbation of the tapping intensity. These authors drive a granular packing to the same packing fraction ρ_0 with three different tapping intensities. Then those three packings are tapped at the same intensity Γ_0 and their behavior depends, for short time, on the previous value of the tapping intensity. These data show a short-time memory effect; the future evolution of the packing fraction depends not only on its initial value but also on its history. Using our experimental setup we recover those memory effects as reported in [20,17]. Do such memory effects also exist at the stationary state ? We can carry out two compaction experiments until the stationary state is reached. For the two experiments we use two different tapping characteristics (i.e. frequency f_1 and tapping amplitude Γ_1 for experiment 1 and frequency f_2 and tapping amplitude Γ_2 for experiment) in such a way that the packing fractions of the two stationary states are equal. Once the stationary state is reached in experiment 1 we can study the response of that packing to taps characterized by Γ_2 and f_2 and compare with the behavior of the packing at stationary state in experiment 2. Since the history of the two packings are different one may expect a transient in the behavior of the packing 1, sign of memory effects. Using this protocol no packing fraction modification can be seen. So no memory effects exist for packings at stationary state. The range of parameters explored remains small but this preliminary result confirms that the packing fraction is an observable that fully characterize the stationary state. Once more, this not true during transient when memory effects [16] reflect the need for, at least, an extra-observable to fully characterize the packing.

7 Conclusions

To Summarize, in this paper we show that granular packings submitted to gentle mechanical taps can reach a stationary configuration. This state does not depend on the initial conditions. A dependence on the duration of the taps is found but our result show a one-to-one correspondence between the final packing fraction and the packing fraction fluctuations. The compactivity, a temperature-like state variable, is then determinated as a function of packing fraction of the stationary state and as a function of the tapping intensity at a given f. We also show that neither memory effects nor annealing are present at stationary state. Our results strongly support the relevance of a fundamental theory of dense granular media.

Acknowledgments : We thank M. Nicodemi, M. Pica Ciamarra and A. Coniglio for useful discussions and N. Taberlet for a careful reading of the manuscript. We acknowledge funding from French ministry of education and research (ACI énergie et conception durable ECD035 - Verres de grains) and from CNRS (PICS 3058 - Relaxation lente dans les milieux granulaires).

References

1. S. F. Edwards and R. B. S. Oakeshott. Theory of powders. *Physica A*, 157(3):1080–1090, 1989.
2. J.B. Knight, C. G. Fandrich, C . N. Lau, H.M. Jaeger, and S.R. Nagel. Density relaxation in a vibrated granular material. *Phys. Rev. E*, 51:3957–3963, 1995.
3. P. Philippe and D. Bideau. Compaction dynamics of a granular medium under vertical tapping. *Europhys. Lett.*, 60(5):677–683, 2002.
4. P. Philippe and D. Bideau. Granular medium under vertical tapping: change of compaction and convection dynamics around the liftoff threshold. *Physical Review Letters*, 91(10):104302, 2003.
5. G. D'Anna, P. Mayor, A. Barrat, V. Loreto, and F. Nori. Observing brownian motion in vibration-fluidized granular media. *Nature*, 424:909–912, 2003.
6. H. A. Makse and J. Kurchan. Testing the thermodynamic approach to granular matter with a numercial model of a decisive experiment. *Nature*, 415:614–617, 2002.

7. A. Barrat, J. Kurchan, V. Loreto, and M. Sellitto. Edwards' measures for powders and glasses. *Physical Review Letters*, 85:5034–5037, 2000.
8. J. Berg, S. Franz, and M. Sellitto. Testing the edwards hypotheses in spin systems under tapping dynamics. *The European Physical Journal B*, 26:349–356, 2002.
9. Martin Depken and Robin Stinchcombe. Fluctuation-dissipation relation and the edwards entropy for a glassy granular compaction model. *Physical Review E (Statistical, Nonlinear, and Soft Matter Physics)*, 71(6):065102, 2005.
10. A. Fierro, M. Nicodemi, and A. Coniglio. Equilibrium distribution of the inherent states and their dynamics in glassy systems and granular media. *Europhys. Lett.*, 59(5):642–647, 2002.
11. Matthias Schröter, Daniel I. Goldman, and Harry L. Swinney. Stationary state volume fluctuations in a granular medium. *Physical Review E (Statistical, Nonlinear, and Soft Matter Physics)*, 71(3):030301, 2005.
12. E.R. Nowak, J.B. Knight, M.L. Povinelli, H.M. Jaeger, and S.R. Nagel. Reversibility and irreversibility in the packing of vibrated granular material. *Powder Technology*, 94:79–83, 1997.
13. G. Lumay and N. Vandewalle. Experimental study of granular compaction dynamics at different scales: Grain mobility, hexagonal domains, and packing fraction. *Physical Review Letters*, 95(2):028002, 2005.
14. Pierre Philippe. *Étude théorique et expérimentale de la densification des milieux granulaires*. PhD thesis, Université Rennes I, 2002.
15. Massimo Pica Ciamarra, Antonio Coniglio, and Mario Nicodemi. Thermodynamics and statistical mechanics of dense granular media. *Physical Review Letters*, 97(15):158001, 2006.
16. C. Josserand, A. V. Tkachenko, D. M. Mueth, and H. M. Jarger. Memory effects in granular materials. *Physical review letters*, 85(17):3632–3635, 2000.
17. P. Richard, P. Ribière, M. Toiya, F. Barbe, R. Delannay, and D. Bideau. *submitted*, 2006.
18. E. R. Nowak, J. B. Knight, E. Ben-Naim, H. M. Jaeger, and S. R. Nagel. Density fluctuations in vibrated granular materials. *Phys. Rev. E*, 57(2):1971–1982, 1998.
19. C. Song, P. Wang, and H.A. Makse. Experimental measurement of an effective temperature for jammed granular materials. *PNAS*, 102:2299–2304, 2005.
20. P. Ribière. *Étude de la compaction d'un milieu granulaire: de l'échelle locale à l'échelle globale*. PhD thesis, Université de Rennes 1, 2005.